Atmospheric
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Discussions

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Interactive comment on "A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of seventeen ice nucleation measurement techniques" *by* N. Hiranuma et al.

Anonymous Referee #1

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General Comments

In the manuscript "A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of seventeen ice nucleation measurement techniques" Hiranuma et al. present an overview of laboratory results from 17 different instruments for immersion freezing experiments using illite NX. Results cover a broad range of temperatures and are compared appropriately to the literature. Both consis-

tencies and discrepancies in the data are discussed in detail, and comparisons are made between methods using dry-dispersed and suspended particles. This work also verifies the efficacy of the IN active surface-site density approach by comparing results within the n_s framework and examining the time-dependence of the immersion freezing process using the SBM. It also examines the dependence of results on freezing mode and presents the main conclusion that temperature change is the major driver of immersion freezing of illite NX particles. The manuscript is well constructed overall and suitable for publication in ACP. However, there are several specific and technical comments below that need to be addressed before submitting the final version.

Specific Comments

Consider subdividing and reorganizing the Introduction into further subsections, e.g. "Background," "Motivation," "Previous work," etc. to improve structural clarity.

Page 22054, Line 5-13: The sentences before "In this study" should be part of the introduction, not in the Methods section.

Page 22055, Line 22-24: The sentence "The influence of dust washing..." seems to suggest that differences in IN propensity were measured for of washed and unwashed particles. However, it seems that in Welti et al., (2014), IC was used to confirm the presence of soluble material, but no experiments were actually performed to test for differences in IN propensity of washed and unwashed samples (rather, these tests are proposed as future work). Please verify that this sentence reflects the actual findings in the cited paper or include references that support this sentence.

Page 22061, Line 15-28: The explanation for why the DLS value for S_{total}/M_{total} is preferred over the TSI-OPS value needs to be clearer and more concise. It might

make intuitive sense to use an SSA value that matches $n_{s,geo}$ for suspension and dry-dispersed measurements, but choosing one over the other need to be better justified. First of all, why is there such a large difference in the S_{total}/M_{total} values reported by DLS and TSI-OPS? Could there be a physical reason that suspended particles (through their interactions with and processing by water) actually have a significantly different S_{total}/M_{total} than dry particles?

Shouldn't the text say 0.49 m^2 g^{-1} is 13 times *smaller* than 6.54 m^2 g^{-1}?

Changing "where the latter..." to "so the latter..." would greatly improve the clarity.

It is unclear whether it is fair to say "$n_{s,BET}$ is especially representative of measurements with suspended samples because minimal corrections..." Yes, the resulting value is based on a relatively simple correction, but how does this make it "especially representative" of measurements if it is strongly dependent on the choice of S_{total}/M_{total}? Granted, the resulting value may indeed be representative of the measurements (though removing the "especially" qualifier would be more appropriate), but a stronger case needs to be made about the appropriate choice for S_{total}/M_{total}.

Since using $n_{m,sus}$ requires an additional assumption than using $n_{m,geo}$, the latter does seem to be a better option, given a better explanation for the choice of S_{total}/M_{total}.

Page 22064, Line 4-15: Could there be aspects of the measurement techniques themselves (or differences in calibrations, corrections, etc.) that could contribute to the differences seen in the SA distributions in Figure 2? How might differences in optical, aerodynamic, and mobility sizing techniques contribute to the differences observed? These (and perhaps other) possibilities should also be mentioned here as potential explanations for the observed differences in addition to possible agglomeration. A more detailed discussion should then appear in Section 4.

Page 22068, Line 10-15: No results are discussed here for CU-RMCS. Include a brief summary here like for the other instruments.

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Page 22075-6, Line 25-1: Is the presence of agglomerates directly measured or just inferred from the results? If the latter is the case, it would be more appropriate for this sentence to say "...may have been carried out in the presence..."

Page 22083, Line 16-17: "agglomerated-fractions based on a relative comparison to D95" implicitly assumes that differences in D_{95} are a result of agglomerations, rather than discussing the possibility of other contributing factors, such as differences in the hydrodynamic size-based, volume equivalent diameter-based, and optical size-based results.

Figure 10: In all other figures, $n_{s,geo}$ is the left column. Please change this figure to match the rest.

Technical Comments

As a general technical comment, the authors are advised to check the consistency of past and present tenses used in the manuscript. Some specific examples are included below, but the flow of the text is sometimes interrupted by unexpected tense changes. Consider using the present tense whenever possible, especially when discussing work done for this study.

Another general technical comment, there are often missing spaces before and after mathematical expressions and symbols. Many are pointed out below, but the authors are advised to verify that all such cases are fixed.

Abstract

Page 22047, Line 27: Consider rewording "Only instruments making measurements with wet suspended samples were able to measure..."

Page 22048, Line 3: Put a space between "to" and "n_s".

Page 22048, Line 9: Remove comma after "spectra"

Page 22048, Line 16: Remove "an"

Page 22048, Line 17: Remove ",thereby,"

Introduction

Page 22049, Line 23: Replace "towards immersion freezing properties" with "for immersion freezing".

Page 22050, Line 3: remove ", which".

Page 22050, Line 6-9: For clarity, change to "Supersaturated conditions with respect to water and ice, as a function of temperature, were created in the simulation chamber vessel by a rapid pressure drop caused by mechanical expansion and subsequent cooling."

Page 22052, Line 28: Change "was" to "is"

Page 22053, Line 6-7: The meaning of "The dataset constitutes a function of..."

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is unclear. Consider rewording as "This dataset captures the functional dependence of... nucleation time on illite NX immersion freezing properties" or something similar.

Page 22053, Line 16: Is the hyphen between parameterization and approach necessary?

Methods

Page 22055, Line 2: Consider using "irregular" rather than "deformed."

Page 22057, Line 12: Replace "about 2" with "~ 2"

Page 22057, Line 21: "is" is inconstant with the tense of the rest of the paragraph.

Page 22058, Line 2: "in the table" should specify the table number.

Page 22059, Line 11-14: As this sentence is currently written, it seems to say that $n_{s,geo}$ represents the geometrically determined surface area (instead of the IN active surface-site density based on geometric size).

Page 22060, Line 7: Consider replacing "under water suspended conditions" with "for experiments using suspended particles."

Page 22060, Line 12: Replace ", therefore S_{total}" with "; therefore, S_{total}.

Page 22060, Line 15-17: Since you are not actually describing a list of steps, consider changing to "...$n_{s,BET}$, the geometric size-based ice nucleating mass, $n_{m,geo}$ (g^{-1}), was first calculated..."

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Page 22060, Line 17: Change S_{total}-to-M_{total} to S_{total}/M_{total} here and throughout for consistency with mathematical notation for the size-selected case.

Page 22061, Line 6: Changing "Lastly" to "Therefore" would provide consistency with the change on Page 22060, Line 15-17.

Page 22061, Line 25: Remove comma after "technique"

Page 22061, Line 27-28: Consider rewording "it is one step further when compared to $n_{s,BET}$ (with an additional assumption of constant size distribution for all suspensions) and two steps further compared to n_m" as "this process requires one more step than when using $n_{s,BET}$ (with an additional assumption of constant size distribution for all suspensions) and two more steps than when using n_m" for clarity.

Page 22062, Line 2-3: Either specify "$n_{s,BET}$ is more representative for suspensions *than*... and $n_{s,geo}$ is better for for dry-dispersed particle measurements *than*..." or simply say something like "$n_{s,BET}$ is suited for suspensions, and $n_{s,geo}$ is suited for dry-dispersed particle measurements."

Results

Page 22062, Line 20-22: Change "wt '%" to "wt %'s" or "abundances" and "was measured" to "were measured."

Page 22063, Line 2: Consider changing "published elsewhere" to "previously published."

Page 22063, Line 17: Change "suggests" to 'suggest."

Page 22063, Line 20-23: For clarity consider rewording, e.g. "Since illite NX particles have significant internal surface area, BET-derived surface areas can be expected to be larger than those derived from the laser diffraction technique. Supporting this notion, ..."

Page 22063, Line 28: Change "These" to "this."

Page 22064, Line 12: Change "discusses" to "discuss."

Page 22065, Line 4: Would be clearer as "$n_s(T)$, (m^{-2} as a function of °C)."

Page 22066, Line 14: Change "500 nm mobility diameter size" to "500 nm mobility diameter size-selected" for consistency.

Page 22066, Line 23-24: Consider changing "with droplets of volume from micro-liter to pico-liter" to "using droplets with volumes in the micro-liter to pico-liter range."

Page 22067, Line 1: Replace "; with the highest temperatures attained" with ". The highest temperatures are attained."

Page 22067, Line 2: Add a comma before "which."

Page 22067, Line 5: "ns(T)" should be written in math mode.

Page 22067, Line 23: Replace "to allow" with "that allows" for consistency.

Page 22068, Line 20-21: Consider rewording "within previously reported uncertainties for immersion freezing experiments" as "for immersion freezing experiments,

within previously reported uncertainties" for clarity.

Page 22069, Line 4-6: Consider changing to "As demonstrated in DeMott et al. (2014), higher RH_w values were required for full expression of immersion freezing in the CFDC. The use of 105 % RH_w in CSU- CFDC does not capture INP activity for many natural dusts, up to a factor of three." for clarity.

Page 22069, Line 14: Remove "available" for clarity.

Page 22070, Line 7: Remove "one"

Page 22071, Line 5-6: It is unclear what is meant by "and, with a slightly better agreement, a time-dependent treatment." Please provide a clearer explanation.

Page 22071, Line 12: Change "from" to "than."

Page 22071, Line 14-15: Why is "(i.e., MRI-DCECC)" included? Also, replace ", which is N_{ice} of" with "of $N_{ice} =$"

Page 22071, Line 17: Change "their" to "the."

Page 22071, Line 20: Replace "therefore" with "so" or use a semicolon to separate the clauses.

Page 22071-2, Line 25-1: Replace "resulting in the data from PINC being in agreement with LACIS..." with "resulting in agreement between the data from PINC and data from LACIS..." for clarity.

Page 22072, Line 9: Replace the comma with a semicolon to separate indepen-

dent clauses.

Page 22073, Line 21: Put a space between "in" and "n_s."

Page 22074, Line 6-8: Consider changing ", whereas" to ". However," and placing a comma before "which" to avoid a run-on sentence.

Page 22074, Line 15: Change "its" to "the."

Page 22075, Line 22: Change "well agreed" to "agreed well."

Page 22075, Line 22: Is "unique" necessary here?

Page 22076, Line 2-4: A space is required before "n_s."

Discussion

Page 22077, Line 21: "axs" should be "axes."

Page 22077, Line 24: Again, "especially" in this context is an unnecessary qualifier?

Page 22080, Line 21: Consider removing "to control of the conditions leading to" for clarity.

Page 22084, Line 13: Replace "be of" with "have."

Page 22085, Line 1: Replace "shows" with "show"

Page 22085, Line 16: Remove "of" for consistency.

Page 22086, Line 11: Commas are unnecessary.

Page 22086, Line 12-14: Consider changing "...PNNL-CIC and IMCA-ZINC both of which measured condensation/immersion and purely immersion mode freezing efficiency of particles, respectively, are in reasonable..." to "...PNNL-CIC and IMCA-ZINC measured condensation/immersion and purely immersion mode freezing efficiency of particles, respectively, and are in reasonable..." for clarity.

Page 22087, Line 6-7: Change "K-feldspar and" and "orthoclase which" to "K-feldspar, and" and "orthoclase, which."

Page 22088, Line 26: Change "the function" to "a function."

Page 22089, Line 8: Change "the function" to "a function."

Page 22089, Line 15: Change "(Garimella et al., 2014)" to "Garimella et al., (2014)."

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